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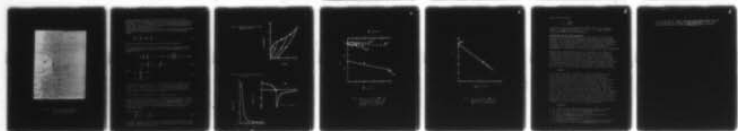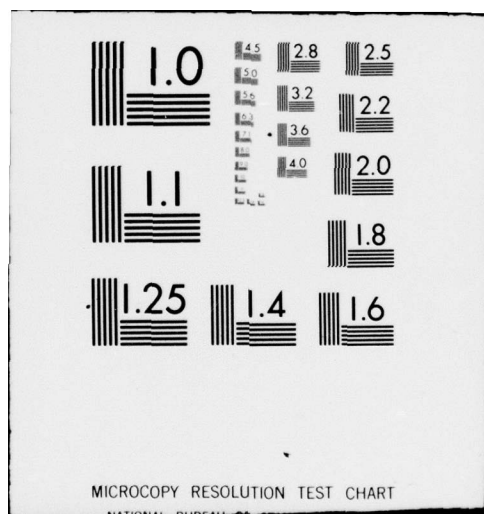   APR 79 E H ANDREWS   DA-ERO-75-G-049
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FRACTURE MECHANICS OF INELASTIC POLYMERS

Final Technical Report

by

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FRACTURE MECHANICS OF INELASTIC POLYMERS

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Abstract - The author's Generalized Theory of Fracture Mechanics
has been applied to several quite different polymeric materials
which have in common a non-linear and inelastic deformation
behaviour. Thus the normal Linear Elastic Fracture Mechanics
cannot be applied. When applied to experimental data the
theory provides a unique characterisation of fracture resistance
and also sheds light on the molecular or microscopic fracture
mechanisms of the material concerned.

INTRODUCTION

Fracture mechanics (FM) is concerned with the propagation of existing cracks
and sets out to define the conditions for such propagation. The first FM
theory was that of Griffth [1] who proposed a criterion for crack growth
(and thus, by implication, fracture of the body) based upon energy
conservation. He said that a pre-existing crack of length c in a stressed
body would propagate if

$$-\frac{d\mathcal{E}}{dA} > \gamma \tag{1}$$

Here the LHS is the 'energy release rate', being the decrease in the
elastic stored energy ($\mathcal{E}$) of the system, with unit area of growth in the
crack surface area (A), and γ is the surface free energy of the solid.
Using linear elasticity theory he was able to evaluate the energy release
rate for a plate containing a short crack under a uniform tensile stress
σ_o. Thus, for plane stress,

$$-\frac{d\mathcal{E}}{dA} = \frac{\pi \sigma_o^2 c}{2E} \tag{2}$$

where E is the Young's modulus. Equating the RHS of equation (2) to γ,
he obtained the critical, or fracture, stress σ_f as,

$$\sigma_f = \sqrt{\frac{2E\gamma}{\pi c}} \tag{3}$$

Following Griffith, other workers recognised that materials were not
generally elastic in the high stress regime around the crack tip, and
suggested that γ be replaced by γ_p, a term including energy losses
dissipated by inelastic deformations in the bulk of the specimen but
notionally assigned to unit area of crack surface. This same approach also
characterises the work of Rivlin and Thomas [2] who used T the 'tearing
energy' instead of $2\gamma_p$.

An alternative fracture mechanics was developed by Irwin [3] who
characterised the intensity of the stress field around a crack by a
stress-intensity factor K such that the stress σ_{ij} at any point $P(r,\theta)$
is given by

$$\sigma_{ij}(P) = \frac{K}{\sqrt{2\pi r}} \, f_{ij}(\theta) \qquad\qquad (4)$$

where f_{ij} are known functions.

Irwin's approach is based on linear elasticity and is commonly referred to as "linear elastic fracture mechanics" (LEFM). We shall use the initials LEFM to embrace any FM theory, including Griffith's, which relies on linear elastic behaviour in the solid.

In LEFM, a small degree of inelastic behaviour can be accommodated by e.g. treating a small plastic zone as an extension of the crack. Large inelastic deformations cannot be treated by LEFM, however.

Since most structural materials are selected for their toughness, they are often neither linear nor elastic. Furthermore they violate the linear elasticity requirement that strains be infinitesimal. For rubberlike solids, Rivlin and Thomas [2] derived equations for the energy release rate or tearing energy T for a variety of specimen geometries without appeal to linear elasticity. Their's is essentially a non-linear elastic analysis. Finally Rice [4] discovered a path-independent contour integral, denoted J, which can be used to characterise the energy release rate in non-linear elastic systems.

Each of these theories is characterized by a fracture parameter which achieves a critical value co-incident with the onset of crack propagation. In Griffith's theory for example this parameter is the energy release rate and its critical value is the solid surface energy γ. Thus, to summarize, we have:-

Theory	Varying parameter	Critical value	Depends on
1. Griffith	$-d\mathcal{E}/dA$ energy release rate	γ	temp. only
2. Irwin	stress intensity factor K	K_c	temp., rate, state of strain
3. Orowan	$-d\mathcal{E}/dA$ or $\tfrac{1}{2}G$	$\tfrac{1}{2}\mathbf{G}_c$	temp., rate, state of strain
4. Rivlin & Thomas	$-d\mathcal{E}/dA$	$\tfrac{1}{2}T$	temp., rate
5. Rice	contour integral J equals $-d\mathcal{E}/dA$	$\tfrac{1}{2}J_c$	temp., rate, state of strain

[N.B. The factor $\tfrac{1}{2}$ appears because A is defined as the area of crack surface which is twice that of crack plane]

Theories 1-3 are based on linear elasticity theory, whilst 4-5 are non-linear elastic theories. None of the theories consider inelastic materials since none consider what happens on unloading the stress field as the crack propagates. They are therefore all concerned with initiation events and can only be used to describe propagation as long as the material is regarded as elastic. Finally, only in Griffith's theory is the critical value parameter explicitly related to the physical properties of the solid.

Turning this around we could say that existing theories suffer from the following deficiencies.

- i) Many assume linear elastic behaviour not found in tough solids, or, usually, in polymeric materials.
- ii) All assume elastic behaviour (i.e. no energy losses).
- iii) Consequently none address themselves to crack propagation which involves unloading of some regions of the stress field and inelastic losses.
- iv) In all theories except Griffith's, the critical parameter which characterises fracture resistance, is purely empirical and not related in any explicit way to the physical or continuum properties of the solid.

The author's "generalized theory of fracture mechanics" was introduced to overcome these various problems. In particular it:-

- i) Copes with non-linear and inelastic materials.
- ii) Concentrates upon the problems of crack propagation, which necessarily involve unloading and mechanical loss.
- iii) Relates the critical fracture parameter explicitly to the continuum properties of the solid.
- iv) Because of its realism in these matters, provides equations which are of predictive value. For example, they predict the critical energy release rate in terms of continuum and physical properties and also predict such phenomena as fatigue and brittle-ductile transitions.

THE GENERALIZED THEORY

The theory has been published elsewhere [5-6], so that we shall simply summarize its results here. These results reside basically in two equations.

Andrews' first equation

The first equation actually represents a family of equations giving the apparent energy release rate in terms of the applied constraints and dimensions of the specimen. These equations take the general form

$$- d\mathcal{E}/dA = k_1(\varepsilon_o, [b_n/\ell]) \; \ell \; W_o \tag{5}$$

Here the LHS is the apparent energy release rate, i.e. the energy release rate that would apply if the material were perfectly elastic. It is thus identifiable with $\frac{1}{2} \mathcal{G}$ for linear solids or $\frac{1}{2}J$ for non-linear elastic materials. On the RHS, $\underline{\ell}$ is a linear dimension and ε_o, W_o are respectively the strain tensor and the input energy density in some identifiable region of the specimen. Usually this region is chosen as one where ε_o, W_o are uniform e.g. regions remote from the crack in simply stressed specimens. The function k_1 depends on ε_o but also upon all non-infinite dimensions b_n of the specimen other than $\underline{\ell}$, normalised by the linear dimension $\underline{\ell}$.

Typically, $\underline{\ell}$ is the crack length if this is short compared with the major dimensions of the specimen. For an edge crack of length $\underline{c}$, for example, and $b_n \gg \underline{c}$

$$- d\mathcal{E}/dA = k_1(\varepsilon_o) \; c \; W_o \tag{6}$$

However, for specimens with crack lengths $c \gg b_1$ where b_1 is one of the major dimensions of the specimen, ℓ would normally be chosen as b_1.

Andrews' second equation

The second equation of the generalized theory is an equation for the __critical__ apparent energy release rate,

$$-(d\mathcal{E}/dA)_c \equiv \mathcal{T} = \mathcal{T}_o \Phi(\dot{c}, T, \varepsilon_o) \tag{7}$$

Here the actual value of $-d\mathcal{E}/dA$ at any phase of crack propagation is denoted by the symbol $\mathcal{T}$, and is equated to the 'surface energy' $\mathcal{T}_o$ multiplied by a 'loss function' Φ.

The 'surface energy' $\mathcal{T}_o$ is more strictly defined as the minimum energy required to break unit area of interatomic bonds across the fracture plane. Its precise physical significance is investigated further in what follows.

The loss function Φ is dependent on the overall state of strain of the body (characterized by ε_o) and also upon any variable of the system which affects the mechanical losses of the material. The most obvious of these variables are the temperature and strain-rate. The latter being controlled largely by the crack velocity $\dot{c}$.

The explicit form of Φ is

$$\Phi = k_1(\varepsilon_o) / \{k_1(\varepsilon_o) - \sum_{PU} \beta g \, \delta v\} \tag{8}$$

where

$$k_1 = (^- \frac{p}{1-p}) \sum_P g \, \delta v$$

- p is a fraction dependent on the curvature of the stress-strain curve of the material. For linear solids $p = 0.5$.

- g is a distribution function of the energy density W throughout the specimen.

- δv is the element of reduced volume, namely the real volume element divided by ℓ^3

- β is the hysteresis ratio (i.e. the fractional energy loss in a stress cycle at a point in the stress field). For elastic solids $\beta = 0$ and for perfectly plastic solids $\beta = 1$.

- P denotes summation over the entire stress field.

- PU denotes summation only over points which unload as the crack propagates i.e. where dW/dc is negative.

The purpose of this report is to detail the progress that has been made in our understanding of the fracture of polymeric solids by application of the generalized theory. In particular we wish to emphasize its value in characterising highly non-linear and inelastic materials and, secondly, in relating their fracture properties to their molecular constitution.

CHARACTERISATION OF NON-LINEAR AND INELASTIC POLYMERS

The value of LEFM for brittle solids lies almost exclusively in its ability to specify a single-valued 'fracture toughness' for a material independent of geometry and loading conditions. For inelastic solids, characterisation has been attempted in terms of J_c and δ_c, but with only partial success. The problem lies in the fact that neither J_c nor δ_c

have explicit physical significance. When, therefore, variations are observed in these quantities it is impossible to say if this is due to a failure of the criterion or to some genuine change in the mechanism of crack growth.

Generalized FM corrects this situation by defining a 'fracture energy' or critical apparent energy release rate, analogous to the $\mathcal{G}_c$ of LEFM. To determine this quantity, $\mathcal{J}$, requires the measurement of a critical loading condition, $W_{o\,crit}$, a dimension (usually the crack length) and the determination of the function $k_1(\varepsilon_o)$. The latter function is conveniently obtained by methods previously described [6] involving load-deflection measurements on specimens containing edge cracks of differing length, c. Referring to Fig. 1, $-\Delta\mathcal{E}$ is given by OAB, OBC etc. and the corresponding ΔA values by c_1, c_2-c_1, etc. times the sheet thickness. Then, from,

$$-d\mathcal{E}/dA = k_1(\varepsilon_o) \; c \; W_o \tag{6}$$

a plot of $-\frac{1}{c}(\Delta\mathcal{E}/\Delta A)$ against W_o is a unique function having the slope $k_1(\varepsilon_o)$ or $k_1(W_o)$ (it is more convenient to use W_o as the variable since ε_o, W_o are in any case uniquely related). The value of W_o is given by area $OAE \div$ specimen volume.

Data for some highly non-linear materials is shown in Figs. 2 and 3. In Fig. 2 are the load-deflection curves for a blown branched polyethylene film (ICI Ltd. XHF film), and in Fig.3 the $k_1(W_o)$ function for this film, for a second PE film (ICI Ltd. XDG film) and for a polyethylene terephthalate film. The shape of the $k_1(W_o)$ is characteristic for yielding solids, having been observed also for highly ductile metals [6], the peak in k_1 occurring at the specimen yield point.

When k_1 is combined with the observed critical values for W_o, and the corresponding crack lengths, a plot of $k_1 W_{o\,crit}$ against reciprocal crack length should give a straight line through the origin having a constant slope $\mathcal{J}$. Such plots are shown in Fig.4 for the PE XHF and the PET films. In this diagram the reciprocal crack length axis has been corrected by a finite-width term, namely the reciprocal of the specimen width, b^{-1}. The $\mathcal{J}$ values obtained are,

PE XHF 4.5 kJ/m^2
PET 5.5 kJ/m^2

We therefore conclude that Generalized FM is capable of producing a geometry-independent fracture parameter for highly non-linear and inelastic materials which is completely analogous to the $\mathcal{G}_c$ of LEFM.

PREDICTION OF FRACTURE ENERGIES

It is an obvious goal of FM to be able to predict critical fracture parameters such as $\mathcal{J}$ from a knowledge of the continuum and molecular properties of solids. It is equally important to understand the physical significance of these parameters in terms of such material properties. These objectives have been brought very much closer by the second equation of Generalized FM.
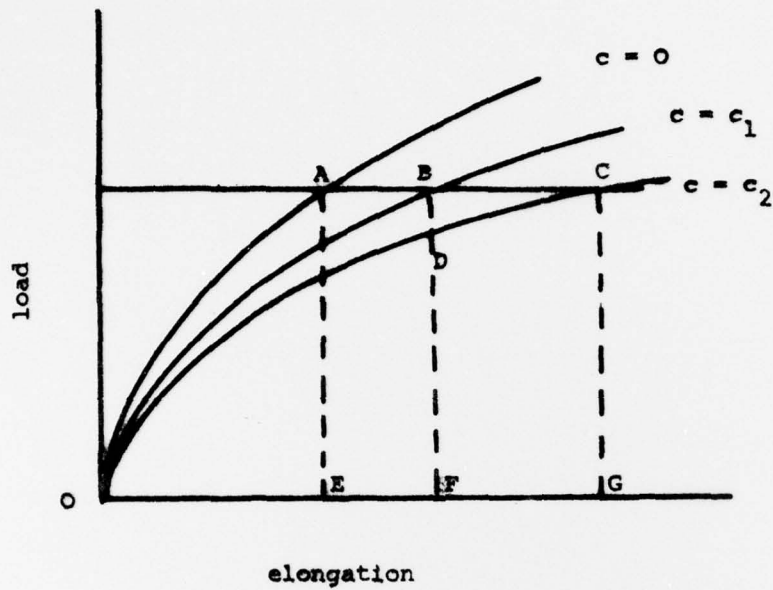
Fig.1. Load deflection curves for single edge notch specimens containing cracks of different length (schematic)
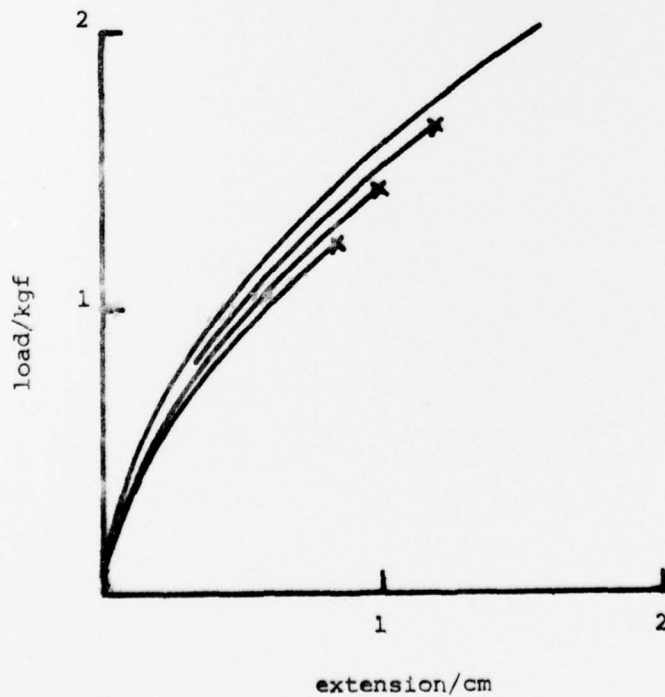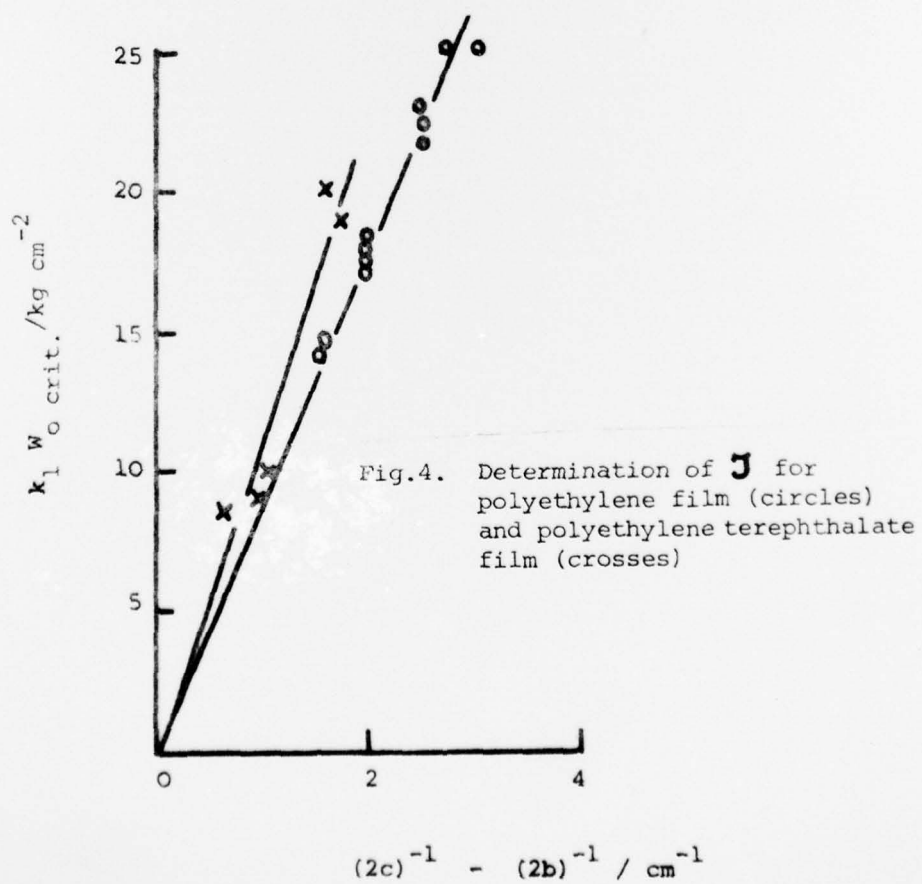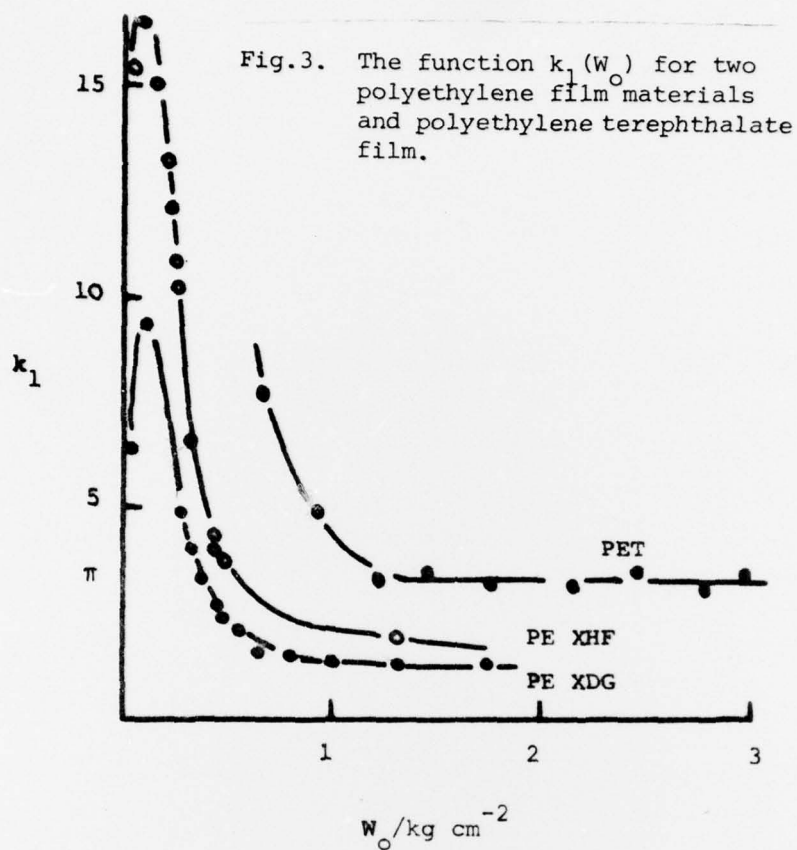

Fig.2. As Fig.1 but showing actual data for polyethylene film. Crosses indicate onset of crack propagation

Fig.3. The function $k_1(W_o)$ for two polyethylene film materials and polyethylene terephthalate film.

W_o/kg cm^{-2}

Fig.4. Determination of $\mathfrak{J}$ for polyethylene film (circles) and polyethylene terephthalate film (crosses)

$(2c)^{-1}$ - $(2b)^{-1}$ / cm^{-1}

Referring back to equation 8, we have

$$\mathcal{J} = \mathcal{J}_o \{\frac{k_1(\varepsilon_o)}{k_1(\varepsilon_o) - \sum_{PU} \beta g \, \delta\nu}\}$$

Clearly, if $\mathcal{J}_o$ and Σ can be evaluated, we have a prediction of the value of $\mathcal{J}$. A complete theoretical solution is still beyond our reach, unfortunately. This is because the energy density distribution function g is not generally known for non-linear deformations. However, g can be determined experimentally from the strain field around a crack, and β, the hysteresis ratio is also experimentally accessible for the general inelastic case. For the special case of an elastic-plastic solid it is sufficient to set $\beta \simeq 1$ in the plastic zone and $\beta = 0$ in the unyielding regions of the specimen.

The calculation of $\mathcal{J}_o$ depends upon the system under study. For elastomeric materials, with a molecular network structure, $\mathcal{J}_o$, can be calculated by the theory of Lake and Thomas (see below). For plastically deforming solids $\mathcal{J}_o$ is expected to be the energy to form and fracture unit area of craze, since any normal evaluation of Σ from a macroscopic strain field would not contain the microscopical craze zone. Below we give examples of both elastomeric solids, including soft polymers like low density PE and plasticized PVC, and of a polymeric glass, polycarbonate.

Elastomers, polyethylene and plasticized PVC
Materials used were SBR and EPDM elastomers, low-density polyethylene and plasticized PVC. Details of these materials are given elsewhere [7].

We have, from equation 8, with suitable abbreviation,

$$\mathcal{J}/\mathcal{J}_o = k_1/(k_1 - \Sigma)$$

$$\mathcal{J}_o/\mathcal{J} = 1 - \Sigma/k_1$$

$$\Sigma/k_1 = 1 - (\mathcal{J}_o/\mathcal{J})$$

Now $\mathcal{J}_o \ll \mathcal{J}$ normally, so that approximately

$$\ln\Sigma = \ln k_1 - (\mathcal{J}_o/\mathcal{J}) \tag{9}$$

This approximation will be very accurate until $\mathcal{J}_o/\mathcal{J}$ increases to about 0.3.

Equation 9 now allows us to check the predictions against experimental values of $\mathcal{J}$ as will be seen below, given a knowledge of Σ, k_1 and $\mathcal{J}_o$.

The parameter $\mathcal{J}_o$ was evaluated by the method of Lake and Thomas [8] using the parameters given in Table 1. Those authors give

$$\mathcal{J}_o = (2/3\pi)^{\frac{1}{2}} \gamma \, \ell \, U \, Nn^{-3/2} \tag{10}$$

TABLE I

Parameters	SBR	EPDM	PE	PVC
$l \times 10^{10}$ (m)	5.15	3.32	2.50	2.50
$m \times 10^{25}$ (N)	12.6	6.79	4.57	10.2
γ	1.54	1.83	1.83	2.77
$U \times 10^{19}$ (J)	12.8	8.78	6.60	6.60
θ (K)	296	296	296	296
η	1.15	1.15	–	–
$\rho \times 10^{-4}$ (N m^{-3})	0.901	0.871	0.922	0.987
$M \times 10^{23}$ (N)	32.7	16.3	–	7.35
$\nu' \times 10^{-25}$ (m^{-3})	1.93	1.93	–	–
$C_1 \times 10$ (MN m^{-2})	2.0	2.0	1.5 ± 0.5	5.5 ± 3.5

TABLE II

Theoretical and Measured Values of the Parameter $\mathcal{T}_o$ (Jm^{-2})

Material	Theoretical	Expt(this work)	Expt(Lake&Lindley)
NR	28	–	40
SBR	34	65 ± 5	60
EPDM	36	65 ± 5	–
p-PVC	29 ± 10	100 ± 50	–
PE	63 ± 11	200 ± 100	–

Notes:- (a) Experimental values always 50% to 100% higher
           than theoretical, both for our own and
           independent data.
       (b) Large error bands occur for non-crosslinked
           materials PE and PVC but even these data are
           consistent with note (a).

where γ is a flexibility parameter, ℓ the length of a monomer unit, U the bond rupture energy for a monomer unit, N the number of network chains in unit volume and $\bar{n}$ the number of monomer units between cross-links. Also

$$N = 2\nu = 2C_1/k\Theta \tag{11}$$

where k is Boltzmann's constant, C_1 the first Mooney-Rivlin constant and Θ the absolute temperature. The cross-link density ν is the sum of chemical links (ν^*) and physical entanglements (ν^1) and its effective value takes account of chain ends thus

$$\nu = (\nu^* + \nu^1)\,(1 - \eta\rho/M\nu^*) \tag{12}$$

where η is a constant, ρ the density and M the average molecular mass of the molecules. In the case of PE and PVC, $\nu^* = 0$ and all the effective "cross-links" are physical in origin.

The results for $\mathcal{S}_0$ are shown in Table II together with data of Lake and Lindley [9]. These authors found that their theoretical values were consistently a factor of about two smaller than experimental values obtained from fatigue experiments and ascribed the difference to deficiencies in the theory.

The evaluation of Σ was carried out as follows. For the case of an edge crack in a semi-infinite sheet, this term takes the form

$$\tfrac{1}{2}\sum_{PU} \beta\,\{x\,\frac{\partial f}{\partial x} + y\,\frac{\partial f}{\partial y}\}\,\delta x\,\delta y \tag{13}$$

where $\beta(x,y)$ is the hysteresis ratio (equal to the fractional energy loss in a stress-strain cycle), x and y are the reduced variables X/c and Y/c respectively and

$$f = W(x,y)/W_0 \tag{14}$$

The summation is taken only over points P which are unloading (PU) i.e. at which the energy density W decreases if the crack propagation.

In order to evaluate Σ we must clearly know β and W as functions of x,y. Since also a propagating crack represents a dynamic situation, and β, W are dependent upon strain rates as well as strain levels, we need to know strain ε and strain rate $\dot{\varepsilon}$ as functions of x,y.

To obtain these data is laborious, but not intrinsically difficult. The experiments carried out to this end are summarized here.
a) An X,Y grid was printed on the sheet containing the edge crack with a mesh size 0.2mm (Plate 1).
b) The crack was caused to propagate by deforming the sheet in an Instron testing machine, the rate of crack growth being controlled by the overall strain applied to the specimen.
c) The strain field surrounding the propagating crack was recorded by photographing the grid.
d) The crack velocity was measured optically.
e) Hysteresis ratio β was obtained for the material as a function of strain level and strain rate by independent simple-extension strain-cycling tests on the Instron.
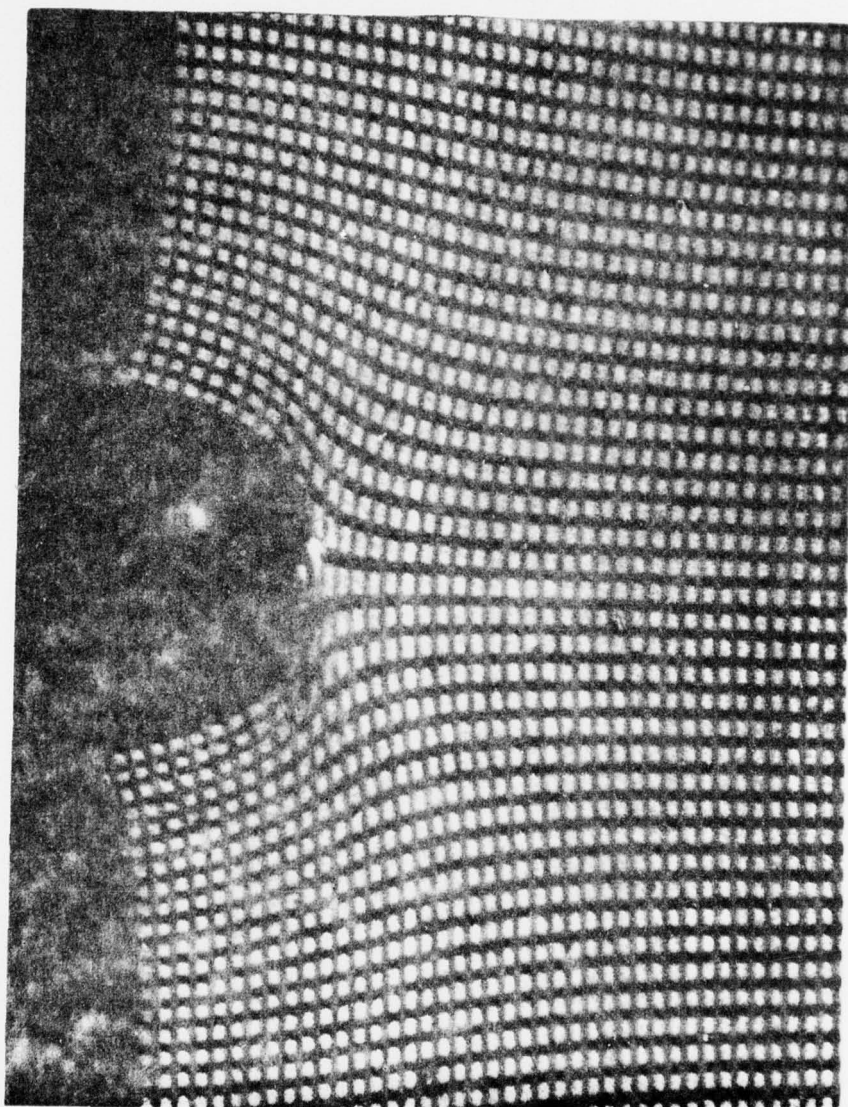
Plate 1 Propagating crack (depth 1mm)
          in a sheet of EPDM rubber,
          showing strain field

Typical hysteresis loops are shown in Figure 5. Ideally, of course, hysteresis data is needed in bi-axial strain states, but this could not be done simply. It is an assumption that at a given temperature and strain rate $\dot{\varepsilon}$, the value of β is uniquely related to the input energy density W regardless of the bi-axiality of strain.

f) The appropriate strain rates at a point P are calculated from the crack velocity and the major principal strain ε_1 at P from the approximate formula

$$\frac{d\varepsilon_1}{dt} = \frac{d\varepsilon_1}{dY} \cdot \frac{dY}{dt} = \frac{d\varepsilon_1}{dY} \dot{c} \tag{15}$$

where the Y axis is the crack axis and $\dot{c}$ the crack velocity.

At this stage we have all the data necessary for the calculation of Σ. The actual evaluation is carried out by the double integration of the argument, thus

$$\Sigma \equiv \frac{1}{c^2} \int_Y \int_{PU} \beta(\varepsilon, \dot{\varepsilon}) X \, dfdY + \frac{1}{c^2} \int_X \int_{PU} \beta(\varepsilon, \dot{\varepsilon}) Y dfdX$$

$$\equiv \frac{1}{W_o c^2} \left\{ \int_Y R_x dY + \int_X R_y dX \right\} \tag{16}$$

where

$$R_x \equiv \int_{PU} h \, X \, dW$$
$$\tag{17}$$
$$R_y \equiv \int_{PU} h \, Y \, dW$$

To evaluate R_x, plots are made of energy density W as a function of the product hX for various values of Y. W is derived from the principal strains ε_1, ε_2 (obtained directly from the distorted grid taking into account that the grid lines are not themselves the principle axes of strain), by use of the Mooney-Rivlin equation.

In all cases X, Y are referred to the undeformed state.

R_x is then obtained by graphically integrating βX with respect to W up to the peak in the W vs X characteristic, since only this region will unload if the crack propagates. R_y is similarly evaluated for various values of X and Σ obtained by graphical integration of the R_x vs Y and R_y vs X curves (see Figures 6 and 7 which represent summation over half the strain field only).

It was found that,

$$\int_X R_y \, dX \gg \int_Y R_x \, dY \tag{18}$$

the second term being negative and of the order of 5% of the first. This simplified subsequent work which neglects the second term and thus halves the effort involved in evaluating Σ.

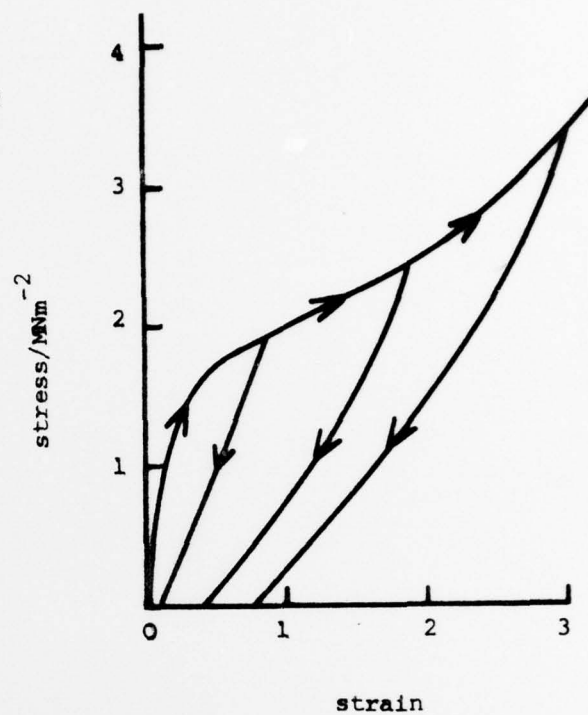Fig.5. Hysteresis loops for EPDM
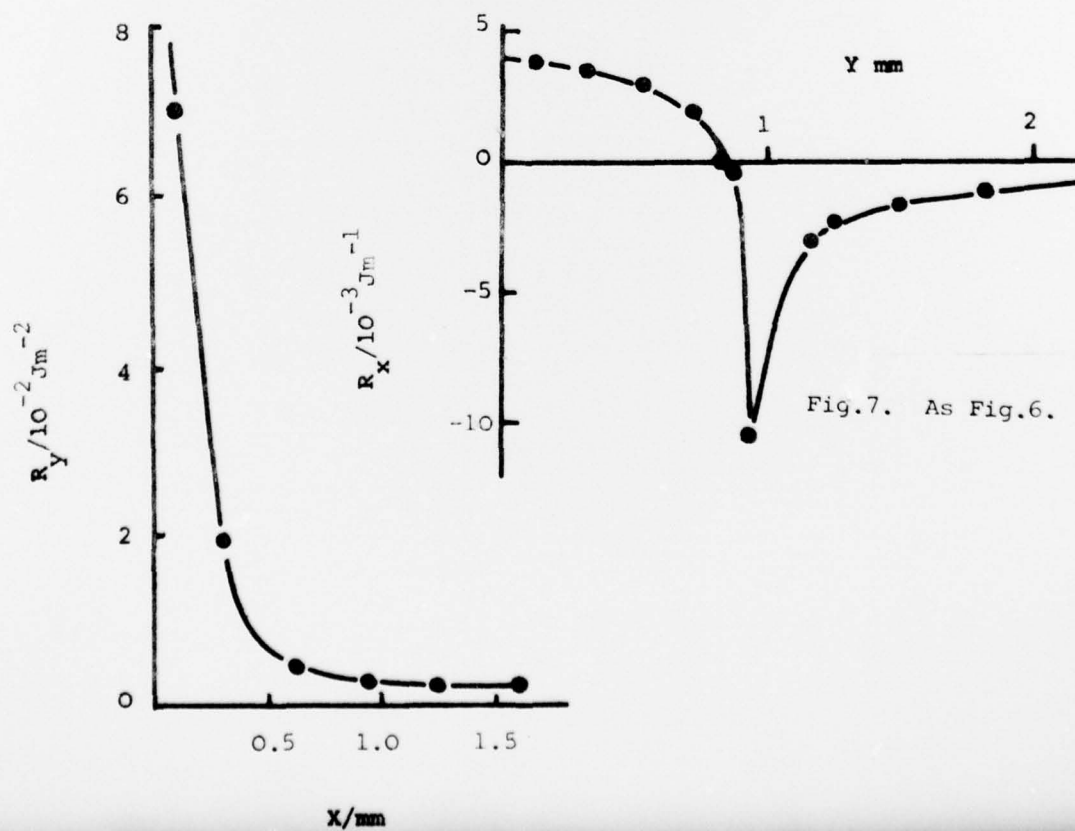rubber at 20°C

Fig.6. A step in the computation of
Σ (see text)

Fig.7. As Fig.6.

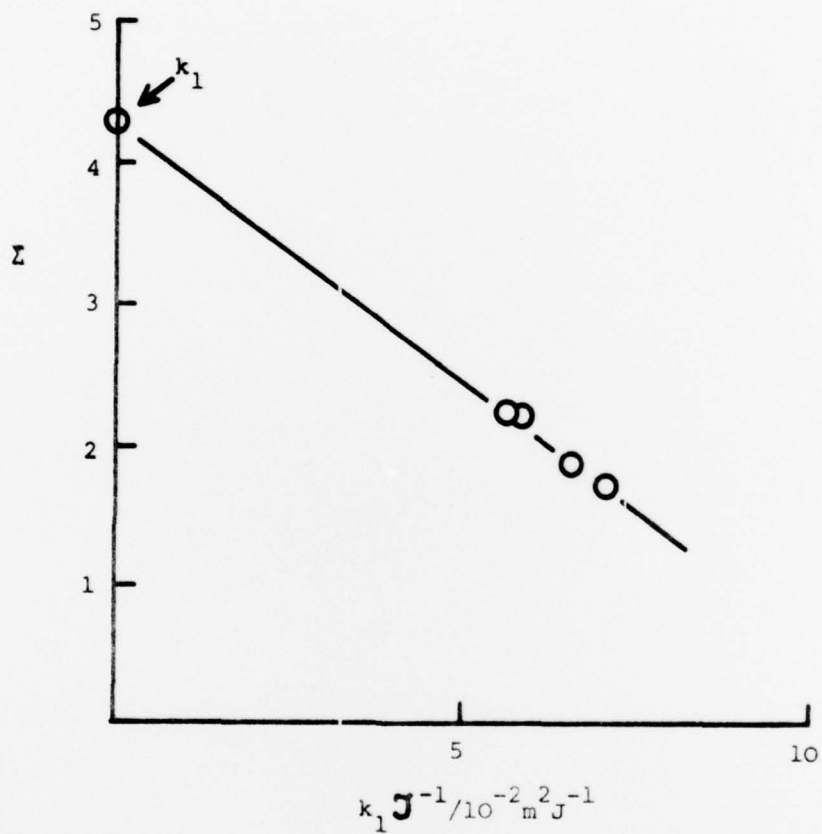Fig.8. Plots of ln Σ versus $\mathbf{J}^{-1}$ for four materials. Linear plots substantiate the theory and give $\mathbf{J}_o$ from the negative slope

Fig.9. Plot of Σ versus $k_1 \mathbf{J}^{-1}$ for polycarbonate. As in Fig.8., $\mathbf{J}_o$ is the negative slope

Using, from equation 8,

$$\Sigma = k_1 - \left(\frac{k_1 \mathcal{J}_o}{\mathcal{J}}\right)$$

we predict a linear relationship between $\mathcal{J}^{-1}$ and Σ with a negative slope of $(-k_1 \mathcal{J}_o)$ and an intercept k_1 at $\mathcal{J}^{-1} \to 0$. Figure 9 shows excellent agreement with this prediction.

Significance of $\mathcal{J}_o$ for polycarbonate

The value of $\mathcal{J}_o$ derived from Fig.9 is large, namely 3.3 kJ/m^2. This compares with 0.2 kJ/m^2 for polyethylene and even less for elastomeric materials. It is obviously impossible to interpret $\mathcal{J}_o$ for polycarbonate in terms of the network theory used earlier to justify high values in elastomeric and similar materials. Even if the entanglement network in polycarbonate is considered to replace the vulcanized network in elastomers, it is difficult to see how such a high figure could be expected.

However, when the $\mathcal{J}_o$ value found here is compared to the $\frac{1}{2} \mathcal{G}_c$ value extrapolated by Ward and co-workers [10] for plane strain (totally brittle) impact fracture in similar materials, a very close agreement is observed. Under these conditions, no plastic flow occurs but only a microscopical craze which precedes the crack. Since in calculating Σ no account was taken of the very high energy density gradients encountered in this microscopical craze zone, it follows that the energy to form the craze must be included in the value of $\mathcal{J}_o$. We conclude therefore that the value 3.3 kJ/m^2 obtained for $\mathcal{J}_o$ is the craze formation energy.

SUMMARY

The fracture and tearing of highly non-linear and inelastic solids, ranging from elastomers, through visco-elastic film materials to elastic-plastic polymer glasses, is well-described by the newly introduced Generalized Theory of Fracture Mechanics. Not only does the theory provide the ability to characterize fracture resistance in non-linear, inelastic materials, but it goes further to provide a new parameter, $\mathcal{J}_o$, which can be related to the mechanism of fracture propagation at a molecular or microscopical level. The interpretation of $\mathcal{J}_o$ varies according to the system under investigation. For the adhesive failure of a Van der Waals bonded interface [11], $\mathcal{J}_o$ is simply half the thermodynamic work of adhesion. For cohesive fracture of network polymers, $\mathcal{J}_o$ is the energy needed per unit area of surface, to break single network chains. For polycarbonate, $\mathcal{J}_o$ is the energy to form the microscopical craze. In all cases the theory allows us to relate the macroscopical fracture mechanics to the microscopical physical processes of energy loss (via β) and failure (via $\mathcal{J}_o$) respectively.

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